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14. ABSTRACT

In this project, we have further developed two-dimensional Fourier transform spectroscopy as a powerful tool to study many-body physics in semiconductor nanostructures. In particular, we investigated the interplay between disorder and Coulomb correlation in semiconductor quantum wells. Monolayer fluctuations in thickness of a semiconductor quantum well lead to the formation of different types of excitons clearly resolvable in the optical spectrum. Whether or not these excitons are coherently coupled via Coulomb interaction is a long-standing debate.

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Report Title

Final Report for ARO grant W911NF-08-1-0348

Development of a Novel Optical Spectroscopy Tool
for Studies of Coulomb Correlations in Semiconductors

ABSTRACT

In this project, we have further developed two-dimensional Fourier transform spectroscopy as a powerful tool to study many-body physics in semiconductor nanostructures. In particular, we investigated the interplay between disorder and Coulomb correlation in semiconductor quantum wells. Monolayer fluctuations in thickness of a semiconductor quantum well lead to the formation of different types of excitons clearly resolvable in the optical spectrum. Whether or not these excitons are coherently coupled via Coulomb interaction is a long-standing debate. Our experiments permit a quantitative measure of the coupling strength between exciton resonances for the first time. In a single, narrow GaAs quantum well, coherent coupling was found to exist between certain exciton resonances but absent between other resonances. Theoretical modeling suggests that strong coherent coupling may be observed only when the Coulomb correlation length is greater than both the disorder correlation and confinement length. Our ultimate goal is to extend 2D spectroscopy to single semiconductor quantum dot molecules (QDMs). As an important step toward this goal, we have characterized a newly developed QDMs produced by molecular beam epitaxial using a hybrid growth method through photoluminescence techniques with high spatial, spectral and temporal resolutions. Our experiments demonstrated that the sizes, shapes, and composition of the QDs within these clusters are highly uniform.

Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

Received		Paper
2011/11/07 1	4	X LI, T ZHANG, S MUKAMEL, R MIRIN, S CUNDIFF. Investigation of electronic coupling in semiconductor double quantum wells using coherent optical two-dimensional Fourier transform spectroscopy, Solid State Communications, (03 2009): 0. doi: 10.1016/j.ssc.2008.12.021
2011/11/07 1	3	Megan Creasey, Xiaoqin Li, J. H. Lee, Zh. M. Wang, G. J. Salamo. Strongly confined excitons in self-assembled InGaAs quantum dot clusters produced by a hybrid growth method, Journal of Applied Physics, (01 2010): 0. doi: 10.1063/1.3369389
2011/11/07 1	2	K. Gundogdu, K. W. Stone, D. B. Turner, X. Li, S. T. Cundiff, K. A. Nelson. Two-Quantum 2D FT Electronic Spectroscopy of Biexcitons in GaAs Quantum Wells, Science, (05 2009): 0. doi: 10.1126/science.1170274
2011/08/13 1	1	Mikhail Erementchouk, Michael N. Leuenberger, Xiaoqin Li. 2d Fourier spectroscopy of disordered quantum wells, physica status solidi (c), (04 2011): 1141. doi: 10.1002/pssc.201000849

TOTAL: **4**

Number of Papers published in peer-reviewed journals:

(b) Papers published in non-peer-reviewed journals (N/A for none)

Received	Paper

TOTAL:

Number of Papers published in non peer-reviewed journals:

(c) Presentations

1. Zheng Sun, Thomas Jarvis, Xiaoqin Li "Interplay between disorder and coulomb correlations in semiconductors", The 4th international conference on Coherent Multidimensional Spectroscopy, Aug 27-30th, Kyoto, Japan, 2008

2. Zheng Sun, Thomas Jarvis, Xiaoqin Li, Mikhail Erementchouk, Michael N. Leuenberger, "Interplay between disorder and coulomb correlations in semiconductors", Quantum Electronics and Laser Science Conference (CLEO/QELS) 2009, Baltimore, MD, June 1-5, 2009.

3. Thomas Jarvis, Zheng Sun, Xiaoqin Li, "A simple implementation of electronic two-dimensional Fourier Transform spectroscopy", Quantum Electronics and Laser Science Conference (CLEO/QELS) 2009, Baltimore, MD, June 1-5, 2009.

4. Zheng Sun, Thomas Jarvis, Xiaoqin Li, Mikhail Erementchouk, Michael N. Leuenberger, "Interplay between Disorder and Coulomb Interaction in Quantum Confined Semiconductors"; Invited presentation at SPIE-photonic west, San Jose, CA, Jan 20th, 2010.

5. Zheng Sun, Thomas Jarvis, Xiaoqin Li, Mikhail Erementchouk, Michael N. Leuenberger, "Quantifying Excitonic Coupling in Disordered Semiconductor Quantum Wells using Electronic 2D Fourier Transform Spectroscopy", APS-March meeting, March 15-19th, 2010.

6. Megan Creasey, J. Bleuse, J. Claudon, N.S. Malik, M. Bazin, I. Maksymov,C. Sauvan, J.-P. Hugonin, P. Lalanne and J.-M. Gérard, Xiaoqin Li, J. H. Lee, Zh. M. Wang, and G. J. Salamo, "Optical Spectroscopy of Single Quantum Dot in Advanced Photonic Structures" PIERS, 2010, Cambridge, MA, July 5th, 2010.

7. Mikhail Erementchouk, Michael N. Leuenberger, Xiaoqin Li, "2d Fourier spectroscopy of disordered quantum wells", Nonlinear Optics and Excitation Kinetics in Semiconductors (NOEKS 2010) in Paderborn, Germany, August 16-19 2010.

8. Yuri D Glinka, Zheng Sun, Xiaoqin Li, Allan Bracker, "Optical Two-dimensional Fourier Transform Spectroscopy of Single GaAs Quantum Wells", APS March Meeting, Dallas, TX, March 23rd, 2011.

9. Thomas Jarvis, Zheng Sun, Xiaoqin Li, Mikhail Erementchouk, Michael N. Leuenberger, "Reflection Geometry Electronic Two-dimensional Fourier Transform Spectroscopy", APS March Meeting, Dallas, TX, March 23rd, 2011.

10. Megan Creasey, Xiaoqin Li, J. H. Lee, Zh. M. Wang, and G. J. Salamo, "Strongly Confined Excitons in Self-Assembled InGaAs Quantum Dot Clusters" APS March Meeting, Dallas, TX, March 23rd, 2011.

Number of Presentations: 10.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received   Paper

     TOTAL:

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received   Paper
2011/11/07 1: 5   Zheng Sun, Thomas W. Jarvis, Xiaoqin Li, Mikhail Erementchouk, Michael N. Leuenberger. Probing many-body interactions in a disordered semiconductor quantum well with electronic two-dimensional Fourier transform spectroscopy, Ultrafast Phenomena in Semiconductors and Nanostructure Materials XIV. , San Francisco, California, USA. : ,

     TOTAL: 1

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):

(d) Manuscripts

Received   Paper
2012/06/18 0: 6   Megan Creasey, Xiaoqin Li,, Ji-Hoon Lee, , Zhiming Wang , Gregory J. Salamo. Self-assembled InGaAs Quantum Dot Molecules with Controlled Spatial and Spectral Properties, Nano Letters (06 2012)

     TOTAL: 1

Number of Manuscripts:

Books

<u>Received</u>   <u>Paper</u>

  TOTAL:

Patents Submitted

Patents Awarded

Awards

The PI has received the following awards in the last five years.

Presidential Early Career Award for Scientists and Engineers (PECASE), 2009

Alfred P. Sloan Research Fellowship   2008-2011

NSF CAREER AWARD 2008

AFOSR YIP AWARD 2008

ONR YIP AWARD 2008

Graduate Students

NAME	PERCENT_SUPPORTED	Discipline
Thomas Jarvis	0.70	
Megan Creasey	0.20	
Kavir Dass	0.20	
FTE Equivalent:	**1.10**	
Total Number:	**3**	

Names of Post Doctorates

NAME	PERCENT_SUPPORTED
Yuri Glinka	0.50
FTE Equivalent:	**0.50**
Total Number:	**1**

Names of Faculty Supported

NAME	PERCENT_SUPPORTED	National Academy Member
Xiaoqin Li	0.08	
FTE Equivalent:	**0.08**	
Total Number:	**1**	

Names of Under Graduate students supported

NAME	PERCENT SUPPORTED

FTE Equivalent:
Total Number:

Student Metrics
This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: 0.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields: 0.00

The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields: 0.00

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale): 0.00

Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering: 0.00

The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense 0.00

The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields: 0.00

Names of Personnel receiving masters degrees

NAME

Total Number:

Names of personnel receiving PHDs

NAME
Thomas Jarvis
Megan Creasey

Total Number: **2**

Names of other research staff

NAME	PERCENT SUPPORTED

FTE Equivalent:
Total Number:

Sub Contractors (DD882)

1 a. Central Florida University   1 b.

                                                                   00000

          Sub Contractor Numbers (c): UTA10-001268
          Patent Clause Number (d-1):
               Patent Date (d-2):
          Work Description (e): The theoretical investigations are going to be performed in close collaboration with Li's group at
     Sub Contract Award Date (f-1): 11/15/2010 12:00:00AM
Sub Contract Est Completion Date(f-2): 2/14/2012 12:00:00AM

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1 a. Central Florida University   1 b.

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Sub Contract Est Completion Date(f-2): 2/14/2012 12:00:00AM

Inventions (DD882)

Scientific Progress

see attachment

Technology Transfer

Final Report for ARO grant W911NF-08-1-0348

Development of a Novel Optical Spectroscopy Tool
for Studies of Coulomb Correlations in Semiconductors

PI: Xiaoqin (Elaine) Li

Physics department, the University of Texas-Austin, Austin, TX, 78712.

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Co-PI: Michael N. Leuenberger
NanoScience Technology Center and Department of Physics,
University of Central Florida

Technical Final Report:

The main focus of the proposal was to develop a new spectroscopy tool, optical two-dimensional Fourier transform spectroscopy, and to investigate electron dynamics in semiconductor nanostructures. This report summarizes our accomplishment during the entire funding period of August 2008-Febuary 2012

A. The importance of the proposed research
Much progress in optical spectroscopy has been made in the last two decades, stimulated by the development of ultrafast laser techniques. Generation and manipulation of short laser pulses on the order of tens of femtoseconds is now a routine. One may speculate that it will be possible to carry out optical spectroscopic measurements as sophisticated as those typical of NMR studies in the future. The development of one advanced technique, optical two dimensional (2D) Fourier transform spectroscopy, is already under way[1, 2]. This spectroscopy technique holds great promise to visualize time-dependent structural changes in molecules[1, 3-8], to follow efficient energy transfer pathways in photosynthesis[9, 10], and to unravel electron couplings and correlations in condensed matter systems[11].

In this project, we have further developed 2D Fourier transform spectroscopy as a powerful tool to study many-body physics in semiconductors. Understanding Coulomb correlations and many-body effects is at the core of condensed matter physics. Optically excited semiconductors are ideal model systems for investigating correlation effects[12, 13]. Following optical excitation, Coulomb interactions lead to the formation of bound electron-hole pairs, known as excitons. Residual interactions between excitons drastically modify the nonlinear optical properties of semiconductors. Therefore, nonlinear optical spectroscopy is a natural choice for investigating correlation effects in semiconductors. In particular, we investigated the interplay between disorder and Coulomb correlation in semiconductor quantum wells. Monolayer fluctuations in thickness of a semiconductor quantum well lead to the formation of different types of excitons clearly resolvable in the optical spectrum. Whether or not these excitons are coherently coupled via Coulomb interaction is a

long-standing debate. Our experiments permit a quantitative measure of the coupling strength between exciton resonances for the first time. In a single, narrow GaAs quantum well, coherent coupling was found to exist between certain exciton resonances but absent between other resonances. Theoretical modeling suggests that strong coherent coupling may be observed only when the Coulomb correlation length is greater than both the disorder correlation and confinement length. Our ultimate goal is to extend 2D spectroscopy to single semiconductor quantum dot molecules (QDMs). As an important step toward this goal, we have characterized a newly developed QDMs produced by molecular beam epitaxial using a hybrid growth method through photoluminescence techniques with high spatial, spectral and temporal resolutions. Our experiments demonstrated that the sizes, shapes, and composition of the QDs within these clusters are highly uniform.

Our research team includes the PI (LI,) three graduate students (Thomas Jarvis, Megan Creasey, and Chandriker Kavir Dass), a postdoctoral researcher, Zheng Sun, and a senior research fellow (Yuri Glinka) and a theoretical group consisting of the co-PI (Michael Leuenberger at the University of Central Florida) and a postdoctoral researcher (Mikhail Erementchouk).

B. Technical and Scientific Results

Experimentally, we have built two experimental set-up of implementing the optical 2D Fourier transform spectroscopy. One set-up is based on a combination of partially collinear geometry and one actively stabilized interferometer. To our knowledge, this is a new development in applying this powerful spectroscopy tool in studies of many-body interactions in semiconductors. Another set-up is based on the standard "box" geometry. We have rebuilt the experiment in the box geometry following the new development in the field[14]. The new set-up has improved stability and allows for phase cycling scheme for improved sensitivity. Using this new and improved set-up, we were able to conduct a systematic study of interplay between disorder and Coulomb correlation in a series of quantum well with different thickness. We observed gradual evolution in 2D spectra consistent with theoretical calculation.

Our simplified 2D experiment is in a partially collinear geometry derived from traditional pump-probe spectroscopy as shown in Fig.1. A pair of pump pulses is generated with variable delay τ and a well-defined phase relationship using an actively stabilized Mach-Zehnder interferometer. A probe pulse, delayed by the mixing time T, is derived from the same laser. The pump pair and probe pulses are focused onto the same spot on a sample. After interacting with the excited sample, the probe pulse is spectrally resolved with a grating spectrometer. The resulting data are imaged with a CCD and saved for subsequent analysis, wherein Fourier transform of these spectral data with respect to τ obtains the two dimensional spectrum.
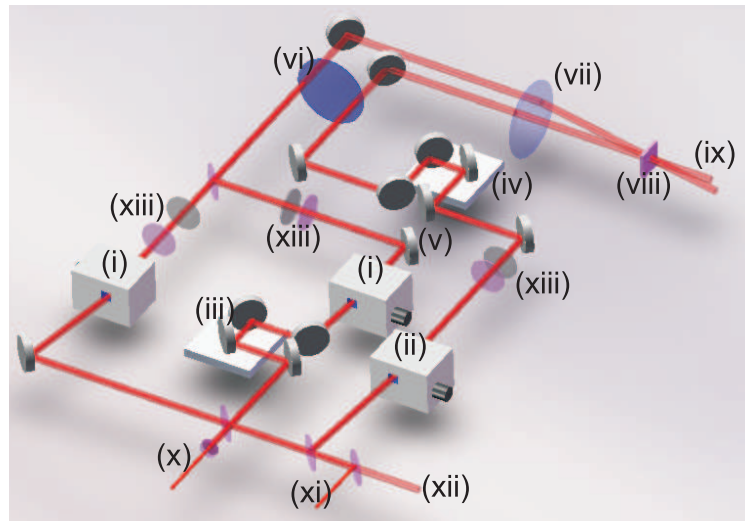
Fig. 1: Experimental schematic. (i) pump AOM's, (ii) probe AOM, (iii) Tau delay stage, (iv) T delay stage, (v) PZT-mounted mirror, (vi) dichroic mirror, (vii) focusing lens, (viii) sample, mounted in liquid He cryostat, (ix) probe beam (to spectrometer), (x) HeNe beam to interferometer photodiode, (xi) HeNe entry point, (xii) Ti:Saph entry point, (xiii) polarization optics (half-wave plate, thin film polarizer).

Our implementation of 2DFTS has several important advantages compared to other such techniques used to study semiconductors. First, this method allows completely flexible control of the polarization configurations. Furthermore, instead of generating a pump-pulse pair using a pulse shaper as in a previous demonstration, we produce the pump pulses with an interferometer; the delay between pump pulses is therefore only limited by the length of the mechanical delay line. This improvement is critical for probing dynamics that exhibit the long time scale decays of interest in semiconductor exciton physics. In addition, our geometry is simpler compared to other actively stabilized 2DFTS methods, as the experiment requires only one

interferometer. Lastly, the experimental arrangement permits easy access for both transmission and reflection measurements.

Using 2D spectroscopy, we investigated the effect of disorder and Coulomb interaction in determining coherent exciton coupling. Disorder at the surfaces and interfaces plays an increasingly important role in ever-shrinking nanostructures. Even in samples of the highest quality, monolayer fluctuations are inevitable. One important type of disorder in quantum wells (QWs) is fluctuations in the QW thickness. A perfect QW can be interrupted by two types of thickness fluctuations, regions that are one monolayer thicker or narrower than the perfect QW. Excitons (bound electron-hole pairs) are fundamental optical excitations in semiconductors. In a narrow QW, exciton wavefunctions may become localized in the presence of disorder. Excitons localized in regions with different QW thickness lead to spectrally distinct resonances.

Whether or not different types of excitons are coherently coupled via Coulomb interactions is an outstanding and much debated question. The presence or absence of coherent coupling among such spatially and spectrally resolved excitons significantly influence energy transfer and photon emission statistics in semiconductor heterostructures such as QWs and quantum wires as well as other systems such as molecular aggregates and photosynthesis. In a broader context, understanding coherent interaction among multiple electronic states is a prerequisite to controlling material properties at the level of electrons, a grand challenge in material science.
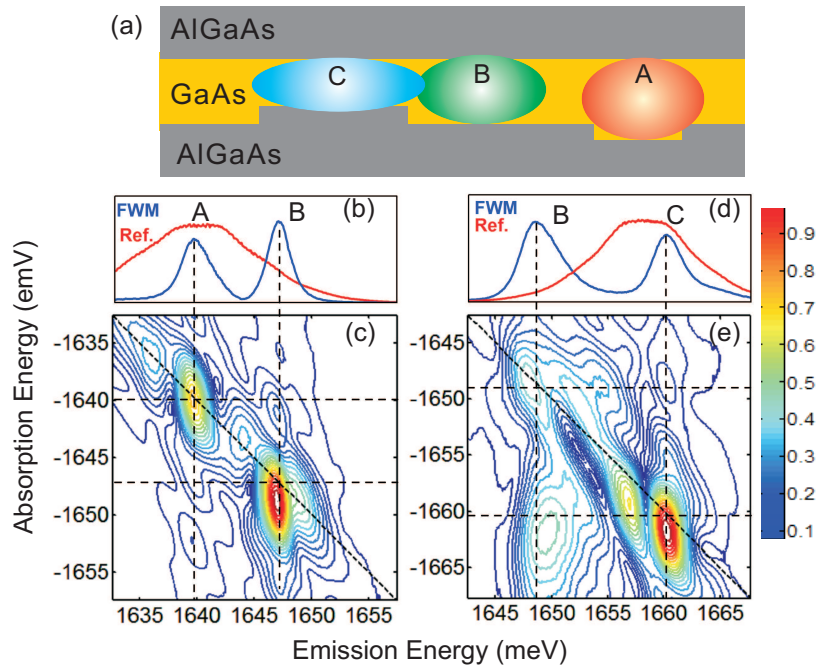

Fig 2: (a) schematics of different types of exciton localized in a narrow QW. (b,d) FWM and (c,e) 2D amplitude spectra when the laser spectrum is tuned to cover different exciton resonances.

In this report, we focus on coherent coupling between different types of excitons in a single GaAS/AlGaAs QW. We first identified three types of excitons confined in different regions of the QW using photoluminescence measurements. We refer to the excitons confined in the wider, average-thickness, and narrower regions of the QW as type A, B, and C excitons, respectively. Using 2DFTS, we were able to separate complex quantum mechanical pathways in the coherent nonlinear response and identify the presence of coherent coupling between type B and C excitons in a narrow QW without any ambiguity. Interestingly, there is no coherent coupling between type A and type B excitons. Theoretical modeling suggests that strong coherent coupling may only be observed when the Coulomb correlation length is greater than both the disorder correlation length and the confinement length.

Our collaborators at the University of Central Florida have focused their effort in on studying 2D Fourier spectra in semiconductor quantum wells (QWs) with spatial inhomogeneities in their thickness. The general dynamics of excitons in inhomogeneous QWs is very complex and still awaits its full description. The special attention, therefore, was paid to specific case of split exciton resonance. The characteristic feature of this case is that exciton states roughly form three classes, according to their contributions to different resonances in the linear absorption spectrum. These states mostly confined to regions where QW thickness supports the respective resonant responses.

The basic description of the 2D spectra of QWs with split exciton resonances as observed in experiments essentially employs the fact that the linear absorption spectrum demonstrates split resonances when the correlation radius of the spatial inhomogeneities exceeds the special confinement length. The physical meaning of this condition is the exciton kinetic energy is smaller than the characteristic value of fluctuations of the local exciton frequencies. Indeed, if inhomogeneities are sufficiently long the the spatial variation of the exciton wave function is smooth implying low value of the kinetic energy. Thus, the fact that the exciton resonance is split justifies using the tight-coupling approximation, i.e. neglecting the exciton kinetic energy, as the first approximation.

Within this approximation the exciton states are characterized by the local resonance frequencies determined by the QW thickness and the Coulomb interaction between excitons confined to different regions of the QW is directly related to existence of the respective cross-resonances in 2D spectra. The Coulomb interaction between excitons has the form of the van der Waals potential, whose characteristic feature is the fast spatial decay. This circumstance is very important because due to the random configuration of the boundaries between the regions with different thickness the Coulomb interaction between the excitons confined to those regions does not imply the formation of the coherent nonlinear response. For latter it is necessary that the nonlinear polarization induced by the Coulomb interaction withstands the effective averaging out over the configuration of the

boundaries. That is, the morphological correlations should decay at shorter spatial scale than that governing turning the exciton-exciton Coulomb interaction into the van der Waals potential.

This imposes quite strict limitations on observing the interaction between the states responsible for the resonant response at different frequencies in the split resonance. On the one hand, the correlations must be sufficiently long in order to ensure the formation of the split resonance. On the other hand, they should not be too long for the Coulomb interaction to be too weak. In thick QWs these two conditions cannot be met because the confinement length in such wells exceeds the exciton Bohr radius and, therefore, the split resonance is supported by inhomogeneities with correlations longer that the characteristic interaction radius. In thin QWs, however, both conditions can be met. The theory developed by the Florida group describes all essential features of the 2D spectra experimentally observed in the series of QWs with different thickness.

Our ultimate goal is to extend the 2D spectroscopy to the limit of studying individual quantum clusters. To this end, we have characterized optical properties of quantum dot molecules on the single dot level using micro-photoluminescence experiments. QDs are often referred to as an artificial atom. The abundance of studies on QDs over the last two decades has been driven by their unique electrical and optical properties. The carriers inside QDs are confined in all three dimensions and thus have a discrete and tunable energy spectrum as well as relatively long carrier lifetimes. These features make QDs the preferred material for a number of optoelectronic devices, such as lasers and photodetectors with enhanced thermal stability, as well as an enticing platform for building novel quantum information processing devices.[1, 2] Further developments in the fabrication of advanced QD structures, like three-dimensional lattices of QDs with controlled spatial and spectral properties, will lead to improved devices such as intermediate-band solar cells.[3]

QDMs, consisting of two or more closely spaced QDs, represent an important step in creating advanced QD configurations.[4] The most frequently studied configuration consists of vertically stacked QDs, which form due to the preferential growth of QDs at the same location as QDs in a buried layer.[5-7] This vertical configuration is attractive because it is relatively easy to grow the QDMs and to vary interactions between QDs by changing the layer spacing. Unfortunately, these QDMs are limited to interactions along one dimension; it is difficult, therefore, to scale beyond two QDs. Alternatively, one may grow QDMs in the same plane, which allows for electrical control and coupling in two-dimensions. Planar QDMs have been grown by a variety of methods such as strain enhanced etching[8-12] and lithography patterning. These methods, however, either involve ex-situ surface preparation or require additional processing steps that add to the complexity of fabrication processes.

Therefore, planar QDMs formed by self-assembly are attractive for their simple growth procedure.

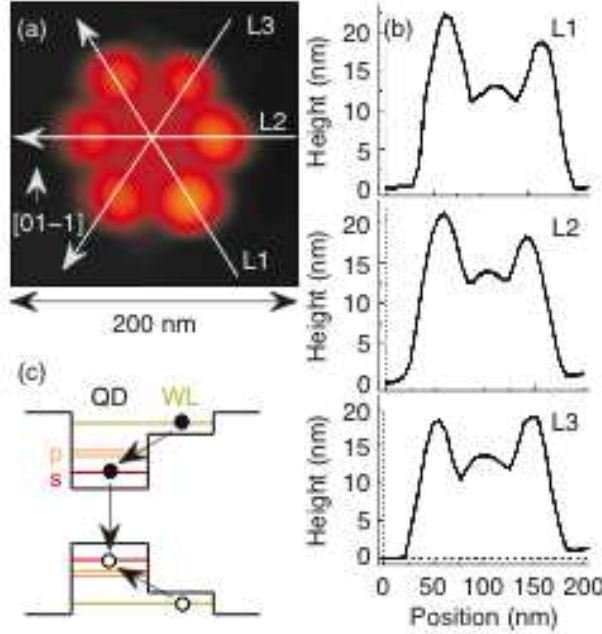We performed the first photoluminescence measurements on individual hexa-QDMs

Fig. 3: (a) AFM image of uncapped QDM. (b) Cross-sections of uncapped QDM along the directions indicated in (a). (c) Energy diagram, including bulk GaAs, InGaAs wetting layer, and QDs.

(Fig. 3) with high spatial, spectral, and temporal resolution. In some QDMs, we observe six exciton transitions close in energy, exhibiting merely a 10 meV spread, with narrow linewidths (< 120 µeV, limited by instrument resolution). The lifetime measurements revealed distinct biexponential decays with approximately a factor of two variation between decay constants for different QDs. The homogeneous exciton energetics and dynamics suggest a high degree of uniformity in the size, shape, and composition of the QDs within the cluster. These QDMs with controlled spatial and spectral properties should enable novel applications based on collective electronic excitations and entangled photon emissions.[18] For example, quantum logic devices built with single QDs are limited to one or two-qubit operations.

We present results of photoluminescence measurements from a particular QDM shown in Fig. 4(a). At low excitation power (8.6 W/cm^2), we observed six closely spaced emission lines at 1267.3 meV (A), 1272.2 meV (B), 1273.3 meV (C), 1274.4 meV (D), 1276.6 meV (E), and 1277.3 meV (F). The very small variation in emission energy suggests that the QDs in this particular QDM have very uniform size, shape, and composition. These transitions all exhibit narrow linewidths ≤ 0.1 meV, corresponding to a long dephasing time. The narrow energy distribution and long dephasing time of QDs within a single cluster make these QDMs particularly attractive for quantum information applications. In previous studies on QD pairs with an average interdot distance of tens of nanometers, applied in-plane electric fields were used to couple exciton transitions separated by a few meV.[9-11] Such controlled coupling between QDs is critical for scalable quantum logic operations or entangled photon generation. In addition, the narrow energy distribution could also be exploited

to create a single photon source continuously tunable over a relatively large wavelength range via a small temperature variation when the hexa-QDMs are placed in photonic nanowires.[44]

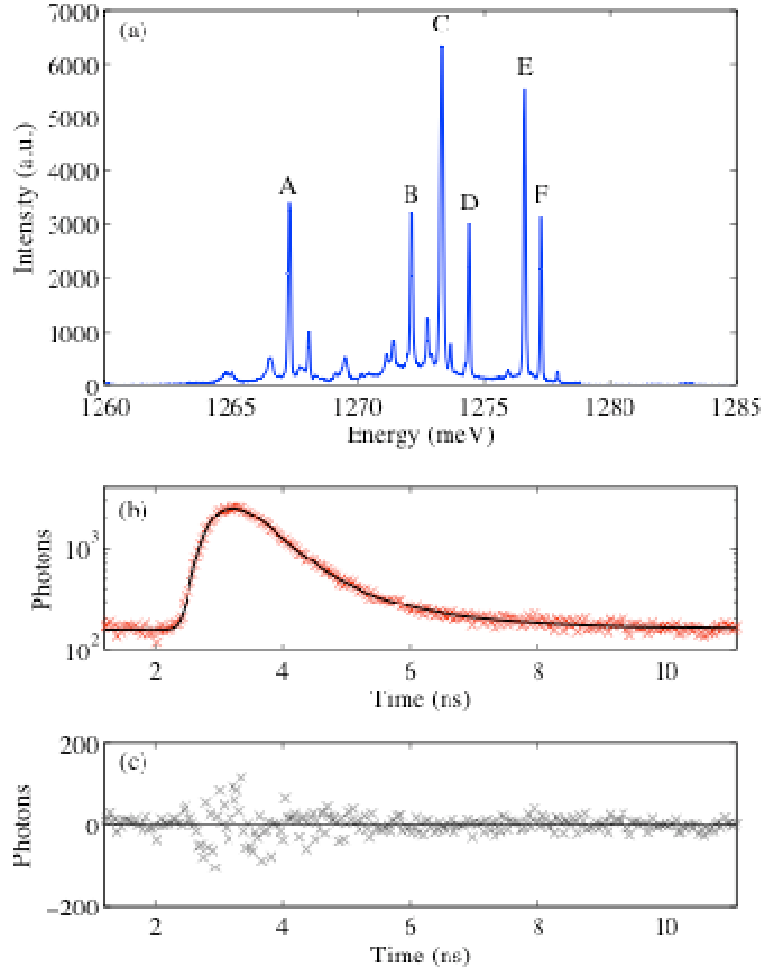We further investigated exciton dynamics of each of the six transitions. An example of the decay dynamics from C, the strongest emission line, is displayed in Fig. 4(b). The TCSPC measurements were made using an excitation intensity of 8.6 W/cm^2, below saturation of the s-shell, to prevent any possible biexciton contribution from complicating the measurement. The decay histograms were fit with three models, a single exponential, a single exponential with a delayed rise time, and a biexponential decay. The biexponential model provided the best fit to the data with a reduced χ^2 value of 1.01, whereas the other models were poorer fits with values of 3.08 and 2.98, respectively. The failure of the delayed rise time model indicates very fast rise times, below the 60 ps resolution of the measurement. This

Figure 4. (a) High resolution μPL of a QDM. Labels correspond to emission energies referenced in the text. (b) TCSPC histogram for emission line A. Raw data is shown as red Xs. Black line shows biexponential fit convolved with system IRF. (c) Residual for biexponential fit to (b). Unpublished results.

suggests that photo-excited carriers are quickly thermalized to the lowest available exciton state. Excitons involved in cascaded processes typically show a delayed rise time due to the finite relaxation time of carriers leaving other states,[39, 45, 46] lending further credibility to our assertion that we are measuring single exciton states without biexciton contributions. The fast rise times also suggest that there is no

carrier transfer between QDs in the clusters on time scales detectable with our experimental setup.

The excellent agreement between the data and the biexponential fit is evident from the random distribution of the normalized residual shown in Fig. 4(c). The chosen biexponential fit is written explicitly as:

$$I(t) = P_0 + \frac{P_f}{\tau_f} e^{-(t-t_0)/\tau_f} \Theta(t-t_0) + \frac{P_s}{\tau_s} e^{-(t-t_0)/\tau_s} \Theta(t-t_0)$$

(1)

where P_0 is the background intensity, t_0 is the initial rise time, $P_{f/s}$ is the intensity of the fast (slow) decay, $\tau_{f/s}$ is the decay time of the fast (slow) signal, and $\Theta(t)$ is the Heaviside step function. Eq. 1 was convolved with a histogram taken of the laser on a reflective surface to correct for the instrument response function of the electronics. The convolved fit was matched to the raw data using a Levenberg-Marquardt algorithm. For this specific resonance, a fast decay of 0.44 ns and a slow decay of 1.70 ns were extracted. All other resonances within this QDM exhibit similar biexponential decays.

A simple two-level model cannot explain the biexponential dynamics. A three-level model, as shown in Fig 4(b), including a non-radiative exciton dark state or trap state is needed to explain this experimental observation. We assume that the radiative (γ_{rad}^b) and nonradiative (γ_{nrad}^b) decay rates of the bright state and the nonradiative (γ_{nrad}^d) decay rate of the dark state are much faster than the bright-dark state transfer rates (γ^{bd} and γ^{db}). As a result, the fast decay rate is determined by $\gamma_f = 1/\tau_f = \gamma_{rad}^b + \gamma_{nrad}^b$ and the slow decay rate is determined by $\gamma_s = 1/\tau_s = \gamma_{nrad}^d$. Biexponential dynamics on similar time scales involving dark excitons have been observed in ensembles of SK InAs QDs.[47] The exact nature of the dark state in our QDMs is unclear without further spectroscopic study, but given that the slow decay component exhibits large variations across the sample in ensemble measurements, we suggest that the dark state may be related to trap or defect states.

In the near future, we intend to investigate possible couplings among individual QDs within the same QDC. Coupling between QDs has only been demonstrated in very limited geometries so far, i.e., between a pair of vertically aligned or planar quantum dots[15-17]. QDCs with complex configurations such as those investigated in this report offer a promising planar geometry for scalable devices. Considering the relatively large spatial separation between individual QDs (tens of nanometers), an external electrical field is likely necessary to induce couplings between the QDs.
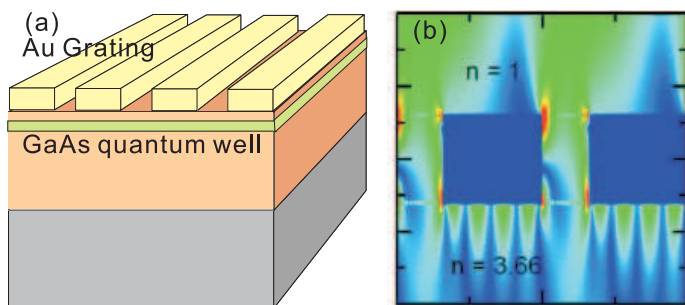
C. Future Directions:

Fig. 5: (a) Schematics of the hybrid sample structure; (b) calculated electrical field distribution of plasmon modes. Figure adapted from Ref 18.

The worked accomplished during this award will motivate us to continue applying 2D spectroscopy to study electronic correlations and energy transfer in semiconductor and hybrid nanostructures. We have already started investigating electronic coupling in a hybrid semiconductor-metal nanostructures, in particular , a GaAs quantum well coupled to a Au grating. Surface plasmon polaritons of the Au grating evanescently couple to excitons in the quantum well (Fig. 5) when the energy and momentum conservations are satisfied under particular excitation conditions. GaAs quantum well structures are widely used in solid state lasers, light emitting diodes, and optical signal modulators and switches in the communication industries. Exploring the feasibilities of building active plasmonic devices which combine the highly confined optical fields of plasmons with the optical gain and nonlinearity of semiconductor quantum wells may lead to the next generation of highly integrated opto-electronics.

The linear optical properties of the hybrid structure have been thoroughly investigated by our collaborators led by Professor Christoph Lienau at the University of Oldenburg[18]. We will investigate the coherent dephasing dynamics and nonlinear response of the coupled electronic resonances in the hybrid structure, which are crucial to predicting properties of potential opto-electronic devices. Specifically, we will investigate the following questions using ultrafast nonlinear spectroscopy methods:

- What is the nonlinear susceptibility of the coupled plasmon-exciton modes? Is it drastically enhanced compared to excitons in bare quantum wells?

- What is the coherent dephasing time for the coupled modes? Does coupling to a quasi-continuum state significantly reduce the exciton coherence time? Or does the energy transfer between two couple modes lead to a dephasing time approximately half of the bare exciton modes?

To answer these questions, we will perform the following spectroscopy measurements:

- Angle resolved pump-probe experiments in the reflection geometry to observe spectral shifts and broadening of coupled exciton-plasmon modes;

- Angle resolved 2DFT experiments to measure modified dephasing dynamics.

It is necessary to conduct these experiments in the reflection geometry. Therefore, the proposed experiments build on the successful development of 2D spectroscopy in the pump-probe geometry during the funding period.

D. Conference Presentation:

1. Zheng Sun, Thomas Jarvis, Xiaoqin Li "Interplay between disorder and coulomb correlations in semiconductors", The 4th international conference on Coherent Multidimensional Spectroscopy, Aug 27-30th, Kyoto, Japan, 2008

2. Zheng Sun, Thomas Jarvis, Xiaoqin Li, Mikhail Erementchouk, Michael N. Leuenberger, "Interplay between disorder and coulomb correlations in semiconductors", Quantum Electronics and Laser Science Conference (CLEO/QELS) 2009, Baltimore, MD, June 1-5, 2009.

3. Thomas Jarvis, Zheng Sun, Xiaoqin Li, "A simple implementation of electronic two-dimensional Fourier Transform spectroscopy", Quantum Electronics and Laser Science Conference (CLEO/QELS) 2009, Baltimore, MD, June 1-5, 2009.

4. Zheng Sun, Thomas Jarvis, Xiaoqin Li, Mikhail Erementchouk, Michael N. Leuenberger, "Interplay between Disorder and Coulomb Interaction in Quantum Confined Semiconductors"; Invited presentation at SPIE-photonic west, San Jose, CA, Jan 20th, 2010.

5. Zheng Sun, Thomas Jarvis, Xiaoqin Li, Mikhail Erementchouk, Michael N. Leuenberger, "Quantifying Excitonic Coupling in Disordered Semiconductor Quantum Wells using Electronic 2D Fourier Transform Spectroscopy", APS-March meeting, March 15-19th, 2010.

6. Megan Creasey, J. Bleuse, J. Claudon, N.S. Malik, M. Bazin, I. Maksymov,C. Sauvan, J.-P. Hugonin, P. Lalanne and J.-M. Gérard, Xiaoqin Li, J. H. Lee, Zh. M. Wang, and G. J. Salamo, "Optical Spectroscopy of Single Quantum Dot in Advanced Photonic Structures" PIERS, 2010, Cambridge, MA, July 5th, 2010.

7. Mikhail Erementchouk, Michael N. Leuenberger, Xiaoqin Li, "2d Fourier spectroscopy of disordered quantum wells", Nonlinear Optics and Excitation Kinetics in Semiconductors (NOEKS 2010) in Paderborn, Germany, August 16-19 2010.

8. Yuri D Glinka, Zheng Sun, Xiaoqin Li, Allan Bracker, "Optical Two-dimensional Fourier Transform Spectroscopy of Single GaAs Quantum Wells", APS March Meeting, Dallas, TX, March 23rd, 2011.

9. Thomas Jarvis, Zheng Sun, Xiaoqin Li, Mikhail Erementchouk, Michael N. Leuenberger, "Reflection Geometry Electronic Two-dimensional Fourier Transform Spectroscopy", APS March Meeting, Dallas, TX, March 23rd, 2011.

10. Megan Creasey, Xiaoqin Li, J. H. Lee, Zh. M. Wang, and G. J. Salamo, "Strongly Confined Excitons in Self-Assembled InGaAs Quantum Dot Clusters" APS March Meeting, Dallas, TX, March 23rd, 2011.

E. Publication in peer-reviewed journals

1. Xiaoqin Li, T. Zhang, R. P. Mirin, Shaul Mukamel, S. T. Cundiff "Investigation of Electronic Coupling in Semiconductor Double Quantum Wells using Coherent Optical Two-dimensional Fourier Transform Spectroscopy", solid state communications, 149, 361-366, 2009.

2. Katherine W Stone, Kenan Gundogdu, Daniel B Turner, Xiaoqin Li, Steven T Cundiff and Keith A Nelson, "Two-quantum 2D Fourire Transform electronic spectroscopy of biexcitons in GaAs Quantum wells", Science, 324, 1169, 2009.

3. Meg Creasey, Xiaoqin Li, J. H. Lee, Zh. M. Wang, G. J. Salamo, "Strongly Confined Excitons in Self-Assembled InGaAs Quantum Dot Clusters Produced by a Hybrid Growth Method", J. Appl. Phys., 107, 104302, 2010.

4. M. Erementchouk, M. N. Leuenberger, X. Li, *2d Fourier spectroscopy of disordered quantum wells*, phys. status solidi (c) **8**, 1141-1144 (2011).

5. Meg Creasey, Xiaoqin Li, J. H. Lee, Zh. M. Wang, G. J. Salamo, "Self-Assembled InGaAs Quantum Molecules with Controlled Spatial and Spectral Properties", Submitted to Nano Letters.

6. Yuri D Glinka, Zheng Sun, Xiaoqin Li, Allan Bracker, Mikhail Erementchouk, Michael N. Leuenberger, "Presence and Absence of Coherent Coupling in Disordered Single Quantum Wells". In preparation.

7. Thomas Jarvis, Zheng Sun, Xiaoqin Li, Mikhail Erementchouk, Michael N. Leuenberger, "A Simple Implementation of Electronic Two-dimensional Fourier Transform Spectroscopy", In preparation.

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